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EMPIRICAL AND SEMI-EMPIRICAL RELATIONSHIPS OF LIQUID-VAPOR, SOLID-VAPOR, AND SOLID-LIQUID SURFACE ENERGIES

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NOMENCLATURE

| | |
|--------------------------|--|
| A | Area |
| A_1, A_2 | Molar area of components 1 & 2. |
| \bar{A} | Average molar surface area |
| a_1, a_2 | Activities of components 1 & 2 |
| a^s_1, a^s_2 | Surface activities of components 1 & 2 |
| B | Constant |
| D_L, D_{ML} | Liquid Diffusion Coefficient at any T and at T_m |
| ΔH | Latent Heat |
| ΔP | Pressure differential across an interface |
| Φ | Contact angle |
| Γ_s | Excess surface solute concentration |
| γ_1, γ_2 | Activity coefficients of components 1 & 2. |
| γ^s_1, γ^s_2 | Activity coefficients of components 1 & |
| H_v | Heat of Vaporization |
| I | Nucleation rate |
| k | Boltzmann's constant |
| N_o | Avogadra's number |
| R | Gas constant |
| R_1 | Radius of curvature |
| R_2 | Radius of curvature |
| σ | Surface energy |
| σ_{lv} | Liquid-vapor surface energy |
| σ_{sl} | Solid-liquid surface energy |
| σ_{sv} | Solid-vapor surface energy |
| T | Temperature |
| T_m | Melting temperature |
| ΔT | Undercooling |
| V | Molar volume |
| W | Work associated with the formation of a surface |
| X_1, X_2 | Atomic compositions of components 1 & 2. |
| x^s_1, x^s_2 | Atomic surface compositions of components 1 & 2. |

BACKGROUND**Surface Energy - Surface Tension Defined.**

The terms surface energy and surface tension are often used interchangeably and have units of J/m^2 and N/m respectively. The choice of terminology is generally dictated by the physical nature of the phenomenon in question. Surface energy is the reversible work need to create a unit area of surface under conditions of constant temperature and volume [1,2].

$$\sigma = \frac{dW}{dA} \quad (1)$$

For solids, surface tension may be thought of as the work required to from a new surface.

Surface energy is associated with the interface existing between two phases. For example, a liquid drop rests on a solid substrate as shown in Figure 1. A liquid-vapor surface energy exists between the liquid and the gas surrounding it. Surface energy exits at the interface formed by the union of two phases, viz., at the liquid-vapor, solid-vapor, and solid-liquid interfaces.

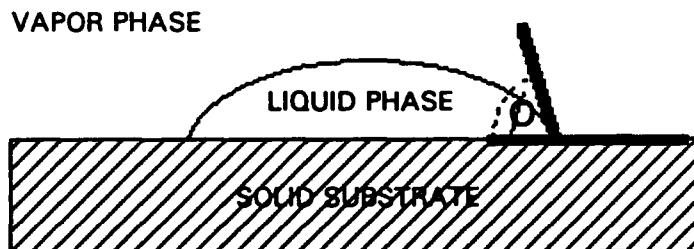


Figure 1 Surface Energy and Contact Angle Established Between Solid, Liquid, and Vapor Phases.

Surface energy may then be considered an interfacial property dependent upon the phases in contact. That is, the

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liquid-vapor surface energy of the droplet in argon will be different than that measured in a vacuum. A summation of forces in the horizontal plane taken at the intersection of the solid, liquid, and vapor phases results in the familiar relationship relating surface energy to contact angle, Eq. 2.

$$\sigma_{sv} = \sigma_{sl} + \sigma_{lv} \cos\phi \quad (2)$$

The liquid is said to wet the substrate if the contact angle is less than 90 degrees.

The curvature of the droplet is associated by the Laplace equation [3,4] to the droplet's liquid-vapor surface energy and the difference between the internal and external pressure of the droplet, Eq. 3.

$$\Delta P = \sigma_{lv} \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \quad (3)$$

The radii of curvature, R₁ and R₂, are orthogonal and tangent to the droplets surface. A spherical drop has a uniform internal pressure. The curvature of a real droplet varies with position, and consequently, so does pressure.

Experimental techniques used to measure surface energy.

A plethora of techniques have been developed to measure surface energy. Iida and Guthrie [3,5] describe several of the most common techniques for liquid metals including (i) capillary rise method, (ii) maximum drop method, (iii) maximum bubble pressure method, (iv) sessile drop method, (v) pendant drop method, (vi) drop weight method, and (vii) oscillating drop method. Other techniques have been developed in order to measure solid-vapor and solid-

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liquid surface tensions [5,6,7] such as (i) zero creep method, (ii) grain boundary grooving, and (iii) surface scratch relaxation. Kumikov and Khokonov [7] report that caution should be exercised in using surface energy data as it is subject to large errors. Methods based upon calorimetry, lattice parameters, and surface scratch relaxation provide results with relative errors of approximately 30-80%. Methods based upon zero creep, contact angles and sessile cylinders have errors of 10-20%. While methods based upon grain boundary grooving and compensated zero creep have relative errors of 1-5%.

Factors Affecting Surface Energy

Temperature, chemistry, and crystallography are just a few of the factors governing surface tension. The surface tension of most elements are reported only at their melting temperature (the influence of temperature discussed in a later section). Alloy chemistry, arguably plays the most significant role in determining the value of surface energy [3,8,&9]. Small amounts of elements such as S, C, N, O, and P which tend to segregate to surfaces can cause a profound change in surface energy. The crystallographic orientation of a solid's surface also influences surface energy. Surface energy is lower for orientations favoring close packed atomic planes because the number of bonds broken is less than on high indices planes.

Surface energy plays a significant role in many material processes e.g., solidification and desulfurization. Sulfur has been associated with the reduced environmental resistance of nickel-based superalloys. The sulfur causes the protective aluminum oxide to spall and greatly reduces the life of coating, e.g., thermal barrier coating. Interestingly, most of the nickel-based superalloys have bulk sulfur contents of less than 12 ppm; however, these

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alloys often exhibit internal and external surface sulfur concentrations in excess of 1,000 ppm. If the surface sulfur is removed either mechanically or chemically, the surface sulfur concentration quickly returns to these high levels upon annealing. The sulfur diffuses to the surface against a high concentration gradient in apparent opposition to Fick's Laws. The explanation, in part, is that sulfur reduces nickel's surface energy and that the chemical potential of sulfur on the surface of nickel is significantly less than in its interior.

Surface active elements such as sulfur, nitrogen, and oxygen are also known to affect the surface tension of other metals [8,10]. The presence of 0.07 wt.% nitrogen in liquid iron reduces its surface surface energy by nearly 15%. As little as 0.09 wt.% oxygen can lower the surface energy of liquid iron by nearly 30% [3], whereas 0.1 wt.% sulfur reduces irons surface energy by nearly 50% [10].

Solidification processes are also greatly affected by a material's surface energy [11]. During casting, how well a molten metal fills a mold cavity is related to both the material's viscosity and surface energy [12]. Metals with lower surface energies require less metallostatic head (pressure) to fill small cavities. The elimination of microporosity from the casting is also related to the liquid-vapor surface energy at the dendritic tips. Since the composition of the alloy at the dendritic tip is constantly changing, so is surface energy.

Most natural solidification processes involve heterogeneous nucleation and surface energy plays a major role in the rate of heterogeneous nucleation. The liquid-solid surface energy play a critical role in determining the nucleation rate of the solid material. Equation (4) describes the rate of heterogeneous nucleation [11].

$$I = B \frac{D}{D_{LM}} \exp \left(-\frac{16\pi\sigma_u^3 T_M^2 V_s^2 f(\theta)}{3\Delta H^2 \Delta T^2 kT} \right) \quad (4)$$

$$f(\phi) = \frac{1}{4}(2 + \cos\phi)(1 - \cos\phi)^2 \quad (5)$$

Nucleation rate is observed to be proportional to the exponential of the liquid-solid surface energy cubed and to $f(\phi)$, a geometric factor. Phi is the contact angle, Figure 1, and is also determined by surface energy, Eq 2.

SURFACE ENERGY COMPILATION, OBSERVATIONS AND CORRELATIONS

There are expressions that enable the estimation of surface energy from a material's fundamental physical and thermodynamic properties. The difficulty in applying these models lies in the lack of accurate materials property data. Therefore, an attempt was made to develop simple, rule-of-thumb type relationships based upon readily obtainable data.

The liquid-vapor, solid-vapor, and solid-liquid surface energies of a number of elements at their melting temperatures were obtained from the literature. Numerous inconsistencies were observed betwixt the various sources. However, where possible, an assessment of the probable accuracy of the data was made and the best data is report in Table 1, [1,3,5,7,11,&13]. An examination of data presented in Table 1, indicates that $\sigma_{sv} > \sigma_{lv} > \sigma_{sl}$. This trend parallels the trend in the entropy change associated with these phase transformations.

Correlation between surface energy and a wide spectrum of properties were investigated; however, heat of vaporization yielded the best results. Figures 2, 3, and 4 illustrate this correlation for the liquid-vapor, solid-vapor, and

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solid-liquid surface energies of various elements. Results of the regression analysis are given in equations 6, 7, and 8.

Table 1. The Liquid-Vapor, Solid-Vapor, and Solid-Liquid Surface Energies of Several Elements at their Melting Temperatures.

| Element | Heat of Vaporization kJ/mol | Liquid-Vapor Surface Energy J/m ² | SOLID-VAPOR SURFACE ENERGY J/m ² | Solid-Liquid Surface Energy J/m ² |
|---------|--------------------------------|--|---|--|
| Ag | 283.79 | 0.912 | 1.162 | 0.126 |
| Al | 329.44 | 0.889 | 1.11 | 0.121 |
| Au | 368.19 | 1.135 | 1.4 | 0.132 |
| Be | 320.17 | 1.39 | 0.95 | |
| Bi | 320.04 | 0.374 | 0.501 | 0.054 |
| Cd | 111.90 | 0.642 | 0.675 | |
| Co | 427.05 | 1.84 | 2.187 | 0.234 |
| Cr | 395.53 | 1.7 | 2.195 | |
| Cu | 335.78 | 1.35 | 1.635 | 0.177 |
| Fe | 413.31 | 1.830 | 1.995 | 0.204 |
| Ga | 271.11 | 0.722 | 0.767 | 0.056 |
| Ge | 371.88 | 0.63 | 0.82 | 0.181 |
| Hg | 60.35 | 0.482 | | 0.024 |
| In | 243.19 | 0.566 | 0.633 | |
| Mg | 145.31 | 0.583 | | |
| Mn | 282.19 | 1.1 | | 0.206 |
| Mo | 656.87 | 2.125 | 2.295 | |
| Nb | 718.56 | 1.93 | 2.235 | |
| Ni | 428.28 | 1.78 | 1.875 | 0.255 |
| Pb | 195.83 | 0.456 | 0.56 | 0.033 |
| Pt | 564.26 | 1.832 | 2.115 | 0.24 |
| Re | | 2.7 | 2.2 | |
| Sb | 264.68 | 0.39 | | 0.101 |
| Sn | 301.52 | 0.554 | 0.676 | 0.059 |
| Ta | 781.38 | 2.13 | 2.49 | |
| Ti | 467.35 | 1.56 | 1.814 | |
| W | 848.50 | 2.42 | 2.67 | |
| Zn | 129.93 | 0.762 | 0.849 | |
| Zr | 607.78 | 1.43 | 1.85 | |

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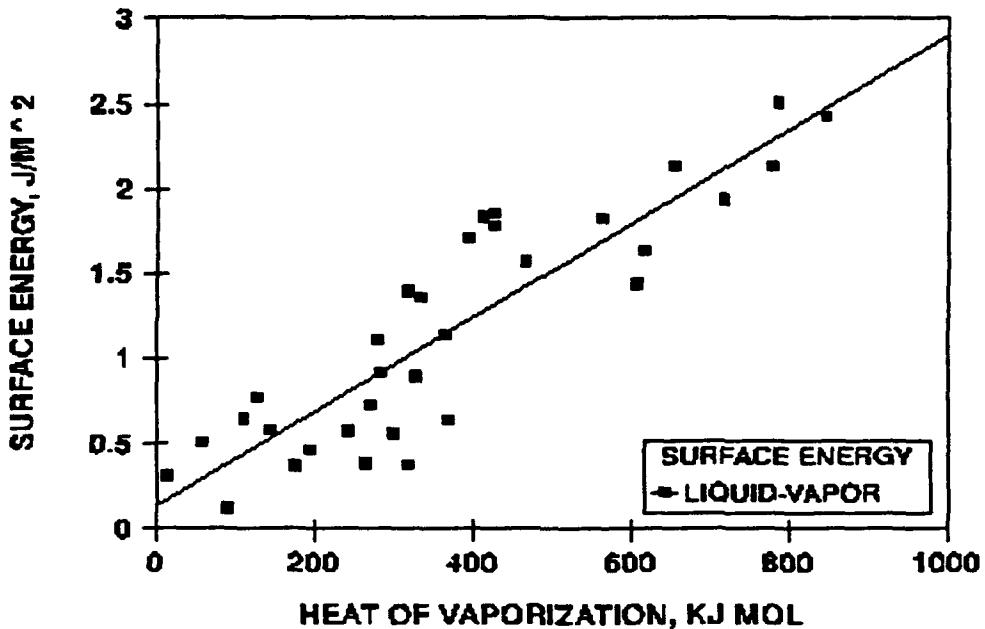


Figure 2 Liquid-Vapor Surface Energy Versus Heat of Vaporization.

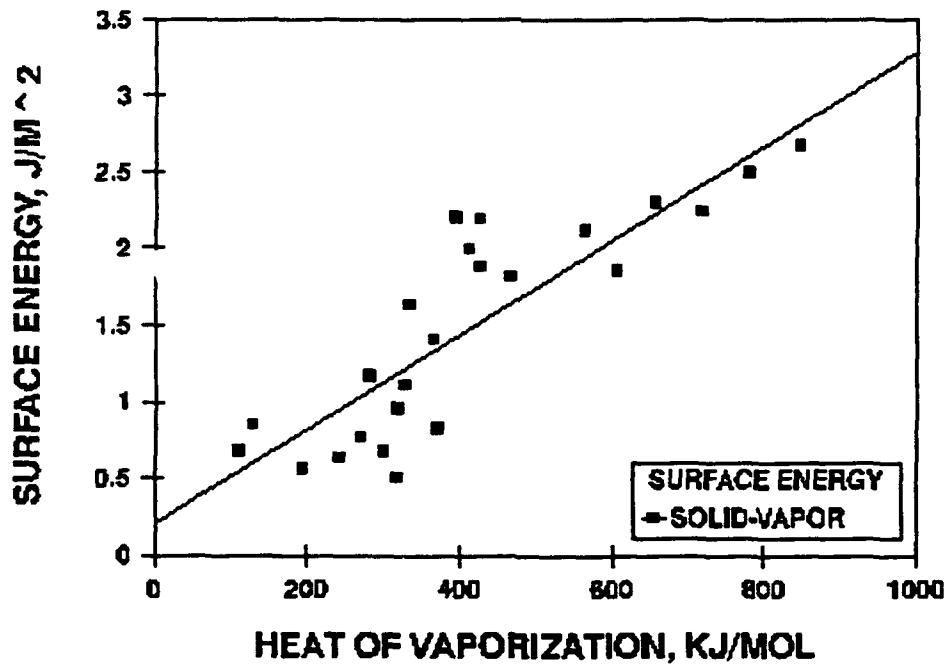


Figure 3 Solid-Vapor Surface Energy Versus Heat of Vaporization.

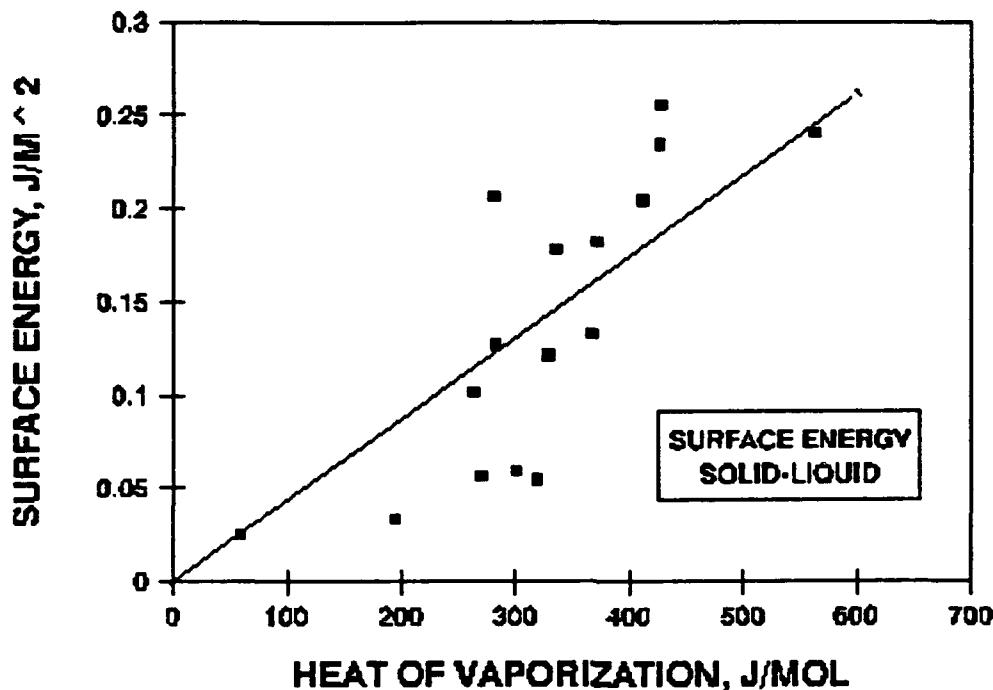


Figure 4 Solid-Liquid Surface Energy Versus Heat of Vaporization.

$$\sigma_{LV} = 2.77 \times 10^{-3} H_V + 0.130 \quad (6)$$

$$\sigma_{SV} = 3.09 \times 10^{-3} H_V + 0.204 \quad (7)$$

$$\sigma_{SL} = 4.36 \times 10^{-4} H_V \quad (8)$$

The ratios of the liquid-vapor / solid-vapor, solid-liquid / solid-vapor, and solid-liquid / liquid-vapor surface energies for the elements listed in Table 1 are presented in Table 2. The data is very consistent as indicated by the standard deviations. Consequently, knowledge of one surface energy allows a reasonable estimation of the other two.

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Table 2. The Ratio of Surface Energies.

| | σ_{lv}/σ_{sv} | σ_{sl}/σ_{sv} | σ_{sl}/σ_{lv} |
|-----|---------------------------|---------------------------|---------------------------|
| AVG | 0.84 | 0.11 | 0.13 |
| STD | 0.04 | 0.02 | 0.02 |

Figure 5 is a plot of surface energy ratios obtained from equations 6, 7, and 8.

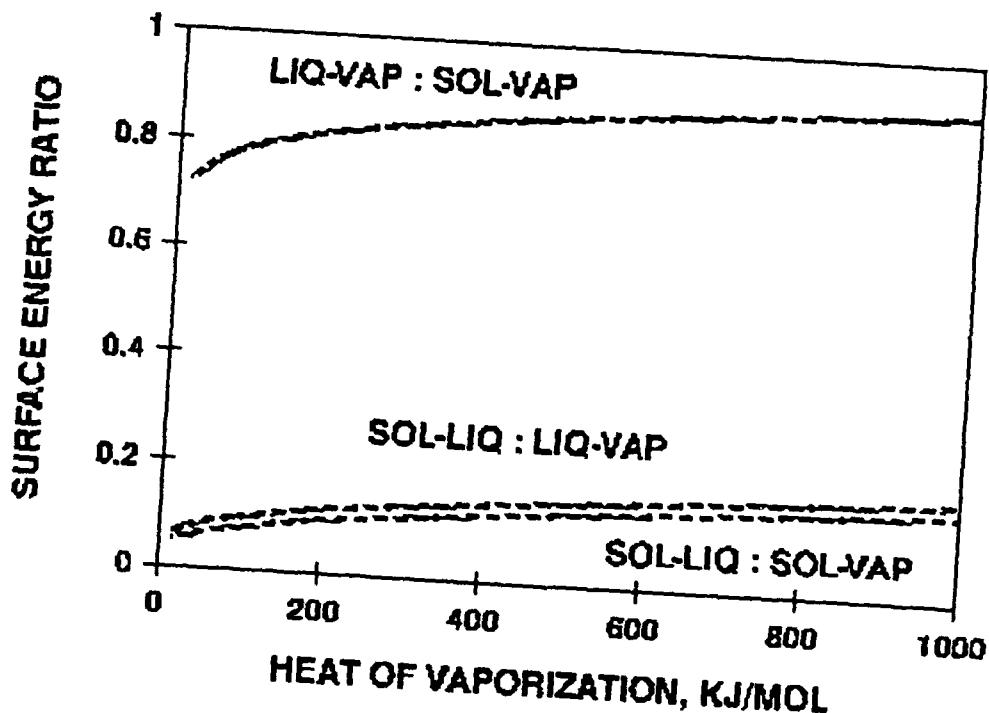


Figure 5 The Ratios of Surface Energy to Heat of Vaporization for Several Elements.

The values are linear over a wide range of heats of vaporization (H_f) from 200-1000 kJ/mol. Deviations from linearity are observed only at low H_f , possibly as the result of the small number of elements with low H_f values.

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used the regression analysis used to generate equations 6, 7, and 8.

The surface energy of a material decreases with temperature at a rate proportional to its entropy. Theoretically, the rate of change of a metal's solid-vapor surface energy has been reported to be $-0.5 \text{ mJ/m}^2\text{K}$ viz a viz the experimentally determined value of $-0.33 \text{ mJ/m}^2\text{K}$ [9].

The liquid-vapor surface energy of a materials also varies with temperature. Surface energy is usually assumed to change linearly with temperature in accordance with equation 9. Linear coefficients of surface energy (LCSE) have been compiled for several elements in Table 3, [3]. While no pragmatic correlations between materials properties and LCSE were found, a statistical assessment of the data was made and is presented in Table 4. The average LCSE is $-0.19 \text{ mJ/m}^2\text{K}$ and varies from -0.05 to $-0.49 \text{ mJ/m}^2\text{K}$. The standard deviation of $-0.13 \text{ mJ/m}^2\text{K}$ suggests that care must be exercised in using values of surface energy obtained using LCSE at temperatures deviating significantly from the materials melting temperature. However, because the LCSE of the material is small relative to its surface energy, the error in using the mean LCSE in lieu of an experimentally determined value is generally small. For example, the surface energy of aluminum 100°C above its melting temperature is $0.854 \text{ J/m}^2\text{K}$; the calculated value using the mean LCSE is $0.87 \text{ J/m}^2\text{K}$, only 1.9% greater.

$$\sigma = \sigma_m + \frac{d\sigma}{dT}(T - T_m) \quad (9)$$

Table 3 The Effect of Temperature on Surface Energy.

| Metal | $-d\sigma/dT$ (mJ/m ² °C) |
|-------|---|
| Ag | 0.19 |
| Al | 0.35 |
| Au | 0.25 |
| Ba | 0.08 |
| Bi | 0.07 |
| Ca | 0.10 |
| Cd | 0.26 |
| Co | 0.49 |
| Cs | 0.06 |
| Cu | 0.23 |
| Fe | 0.49 |
| Ga | 0.10 |
| Ge | 0.26 |
| Hg | 0.20 |
| In | 0.09 |
| K | 0.08 |
| Li | 0.14 |
| Mg | 0.35 |
| Mn | 0.20 |
| Na | 0.10 |
| Ni | 0.38 |
| Pb | 0.13 |
| Rb | 0.06 |
| Sb | 0.05 |
| Sn | 0.09 |
| Ti | 0.26 |
| Tl | 0.08 |
| U | 0.14 |
| Zn | 0.17 |

Table 4 $d\sigma/dT$ Statistics.

| Property | Value |
|--------------------|-------|
| Mean | 0.19 |
| Median | 0.14 |
| Standard Deviation | 0.13 |
| Variance | 0.016 |
| Minimum | 0.05 |
| Maximum | 0.49 |

METAL ALLOYS

This paper has focused on the examination of surface energy of pure elements because of the lack of credible data for most alloy systems. However, materials of engineering significance are typically multicomponent alloys. The surface tension of many alloys can be estimated using classical thermodynamics [1,2,3,14,15,&16]. The thermodynamics of two types of systems will be considered:

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- (i) a dilute alloys in which Henry's law is obeyed, and
- (ii) an ideal solution for which Roult's law is applicable.

For dilute alloys, the Gibb's isotherm, equation 10, may be used in order to determine the surface energy or conversely excess surface concentration of solute. Unfortunately, the application of the model requires the empirical determination of either the surface energy or surface composition. The plot of the change in surface energy per change in the log of composition is proportional to excess surface energy.

$$\Gamma_s = \frac{d\sigma}{RTd(\ln X^s)} \quad (10)$$

A simply applied model for the surface tension of an ideal binary mixture was developed by Guggenheim [17]. Benard and Lupis [4] determine the surface tension of silver-gold alloys and demonstrated the applicability of the Guggenheim model to that system.

$$\exp\left(\frac{-\sigma\bar{A}}{RT}\right) = X_1 \exp\left(\frac{-\sigma_1\bar{A}}{RT}\right) + X_2 \exp\left(\frac{-\sigma_2\bar{A}}{RT}\right) \quad (11)$$

Equation 11 represents fairly well the behavior of many systems such as the Cu-Ni, Au-Bi, Be-Bi, Pb-Sn etc. [4]. Unfortunately, for systems which deviate substantially from ideality, the performance of Guggenheim's model has been rather poor, especially for solid solutions [3].

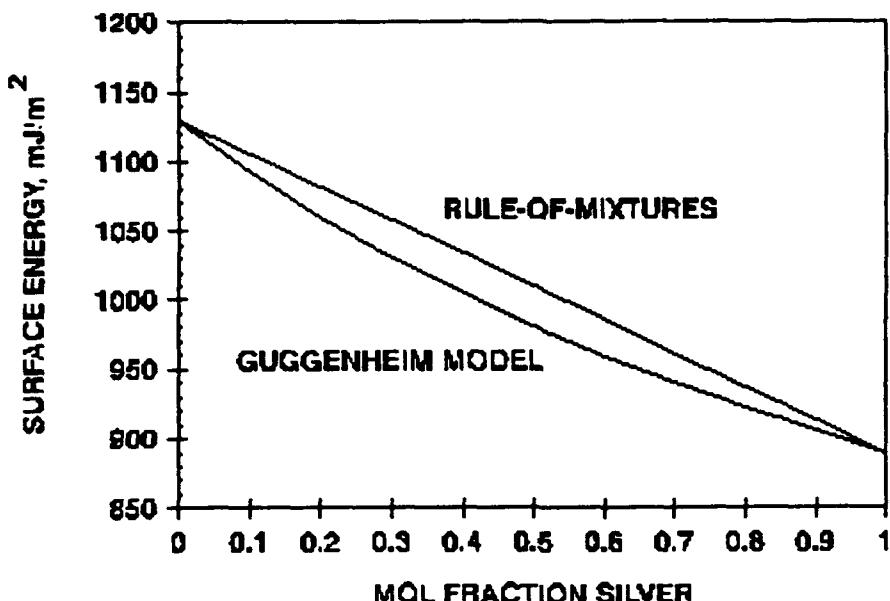


Figure 6 Calculated Surface Energy for the Silver-Gold System.

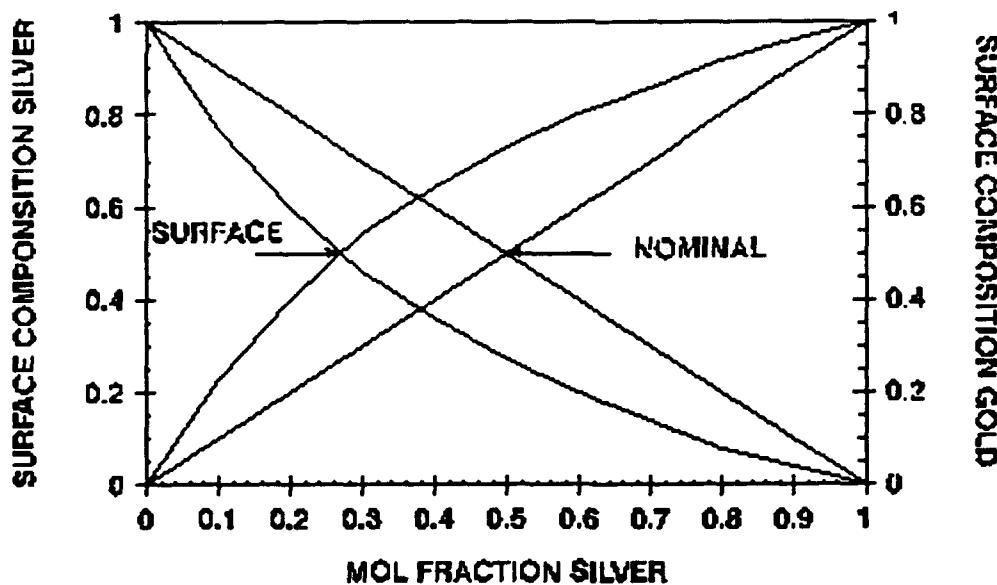


Figure 7 The Nominal and Calculated Surface Compositions for Silver-Gold Alloys.

Using data from Bernard and Lupis [4], figure 6 was generated using the rule-of-mixtures and equation 10 for an ideal solution. The surface composition of the alloy system

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was calculated from equations 12 and 11 [15] and is presented in Figure 7.

$$\sigma = \sigma_1 + \frac{RT}{A_1} \cdot \ln \frac{a_1^s}{a_1} \quad (12)$$

$$A_1 = 1.09 N_o^{1/3} V_1^{2/3} \quad (13)$$

Figure 7 illustrates an important and generally applicable rule: elements with the lower bulk surface energy segregate to the surface. For example, a silver-gold alloy with a nominal bulk composition of 0.5 atom % silver and 0.5 atom % gold has a surface composition of 0.73 atom % silver and 27 atom % gold.

SUMMARY AND CONCLUSIONS

An extensive compilation of surface energy data for elements at their melting temperature has been presented, Table 1. The data was analyzed in order to determine easy to use, rule-of-thumb type relationships. It was found that the liquid-vapor, solid-vapor, and solid-liquid surface energies of an element varied linearly with their heats of vaporization, Equations 6, 7, & 8. The ratios of the elements' liquid-vapor / solid-vapor, solid-liquid / solid-vapor, and solid-liquid / liquid-vapor surface energies are remarkably consistent and are therefore useful in estimating one from another. Temperature's affect on surface energy was observed to be minimal; and therefore, estimations made using the average LCSE are not likely to be significantly in error.

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The data for the surface energy of alloys is not nearly as available and reliable as it is for the elements. The surface energy of alloys with even minute amounts of surface active elements (C, S, O, P, N, etc.) vary greatly from ideality or "the rule-of-mixtures". These systems were best described using Gibb's isotherm and empirical data. For alloys which don't deviate far from ideality, the Guggenheim model can be used to estimate surface energy. This data enables the calculation of the excess surface solute concentration. In a multicomponent alloy, the element with the lowest surface energy tends to segregate to the alloy's surface.

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